

# PATENT ABSTRACTS OF JAPAN

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(54) MANUFACTURE OF SILICONE COMPOSITE BODY

(57)Abstract:

**PURPOSE:** To easily manufacture silicone composite body consisting of surface layer and base layer by a method wherein layer made of silicone composition, which turns into the form of gel by hardening, and silicone composition, which is provided on the surface of the layer and turns into the form of elastomer or the like, are hardened simultaneously.

**CONSTITUTION:** The silicone composite body concerned consists of base layer made of silicone gel and surface layer, which is formed on the surface of the base layer and made of silicone elastomer or silicone resin. In this case, at the manufacturing of the silicone composite body, firstly, layer made of hardening silicone composition, which hardens into gel-like hardened matter, is provided. Next, on the surface of the above-mentioned layer, layer made of hardening silicone composition, the specific gravity of which is smaller than that of the above-mentioned composition and which hardens into elastomer-like or resin-like hardened matter. In succession, the respective layers made of the above-mentioned compositions are hardened simultaneously. Thus, silicone composite body consisting of surface layer having high hardness and the like and base layer having low hardness and the like is easily manufactured by single hardening treatment.

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## DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Industrial Application]This invention relates to the manufacturing method of the silicone complex which consists of base phase which consists of silicon gels, and a surface layer which consists of the elastomer or resin formed on this phase.

[0002]

[Description of the Prior Art]Since the gel hardened material of silicone is called what is called silicon gel and it has the outstanding characteristics, such as the stability of electric insulation and an electrical property, and pliability, As the electrical and electric equipment, potting of electronic parts, and an object for closure, especially, circuit elements for control, such as a power transistor, IC, and a capacitor, are covered, and it is used as covering material for thermal and protecting from a mechanical obstacle.

[0003]However, since the surface of silicon gel has strong adhesiveness, garbage and dust adhere easily and there is a problem on the work of a molded product \*\*\*\*\*ing and not being made on silicon gel. When removing the garbage etc. which adhered since the mechanical strength was insufficient, there is a problem on which silicon gel suffers a loss. Methods of canceling a fault which was mentioned above include the method of providing a surface protection layer in the silicon gel surface so that it may state below.

[0004](1) How to provide the protective layer of other organic system high hardness resin on the surface of silicon gel, [0005](2) How to carry out heat cure and to provide the silicone layer of the shape of an elastomer, or the shape of resin in the surface after giving the ORGANO hydrogen polysiloxane etc. on the surface of silicon gel and diffusing a gel surface as indicated to JP,1-25704,B, [0006](3) As indicated to JP,61-277414,A, The ORGANO hydrogen polysiloxane is given to the surface of the silicone gel composition of an uncured state, How to carry out simultaneous hardening of the surface layer from which it heated and an inside silicone gel composition and the ORGANO hydrogen polysiloxane were incorporated before being spread, and to provide a silicone elastomer layer on the surface of a silicone gel composition, [0007](4) The method of giving the constituent which becomes a silicone elastomer layer or a silicone resin layer by hardening to the surface of the hardened silicon gel, making it harden it, and providing a surface layer in it, etc. are known.

[0008]

[Problem(s) to be Solved by the Invention]However, in the method of the aforementioned (1), since layer indirect arrival power is insufficient, it is easy to generate interlaminar peeling. So that it may not remain in the method of (2), not having hardened the ORGANO hydrogen polysiloxane on the silicon gel surface, The kind

of ORGANO hydrogen polysiloxane with the residual functional group in silicon gel and the functional group which reacts, and quantity must be chosen, or it is necessary to wash or wipe away after hardening. In the method of (3), since a silicone gel composition is an uncured state when heating, the ORGANO hydrogen polysiloxane given to the surface of this constituent is involved in by the convection into a silicone gel composition. As a result, a surface layer hardens unevenly, and a good protective layer is not obtained, and also control of the thickness of a surface protection layer and hardness is difficult. In order to harden the constituent which becomes a silicone elastomer layer or a silicone resin layer in the method of (4) after hardening a silicone gel composition, a process becomes complicated and, also in time, is disadvantageous.

[0009]Then, SUBJECT of this invention is providing the manufacturing method which can manufacture easily the silicone complex which consists of base phase which consists of silicon gels, and a surface-protection phase which has this base phase and good adhesion and was made in a reliable silicone elastomer or silicone resin at an easy process.

[0010]

[Means for Solving the Problem]That is, this invention is a manufacturing method of a silicone complex which consists of a surface layer which consists of a silicone elastomer or silicone resin formed in base phase which consists of silicon gels, and its surface, [0011](a) It averages among all the organic groups combined with a silicon atom contained in one molecule. More than 0.025 mol % is an alkenyl group, And an alkenyl group which has the viscosity at 25 °C in the range of 10 - 100,000 cP and which was combined with a silicon atom in one molecule is averaged. Organopolysiloxane which it has 0.1-2 pieces, [0012](b) It is the ORGANO hydrogen polysiloxane which has the hydrogen atom combined with a silicon atom in [ 1-50 ] one molecule, per [ which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a) / ] alkenyl group -- what is blended so that it may become 0.3-2.0 individual -- and [0013](c) On a process of providing a phase which consists of a hardenability silicone composition (A) containing an addition reaction catalyst which hardens and generates silicon gel, next a phase of said constituent (A), [0014](d) inside 0.05 [ an average of ] of all the organic groups combined with a silicon atom contained in one molecule -- more than mol % being an alkenyl group, and, And organopolysiloxane which has the viscosity at 25 °C in the range of 10 - 100,000 cP and which has two or more alkenyl groups combined with a silicon atom in one molecule, [0015] (e) It is a hydrogen atom combined with a silicon atom in one molecule 5-100 It is the ORGANO hydrogen polysiloxane which \*\*\*\*, per [ which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a) / ] alkenyl group -- what is blended so that it may become 0.3-2.0 individual -- and [0016](f) Contain an addition reaction catalyst and specific gravity is smaller than said constituent (A), A manufacturing method which has the process of presenting hardening with a layer which consists of a process of providing a layer which consists of a hardenability silicone composition (B) which hardens and turns into a hardened material of the shape of an elastomer or the shape of resin, a phase which consists of said constituent (A), and a constituent (B) simultaneously is provided.

[0017]A manufacturing method of this invention presents hardening with what laminated a layer which consists of a silicone composition (B) which has small specific gravity relatively on the surface of a phase which consists of a silicone composition (A) which has big specific gravity relatively. A surface layer of a constituent (B) in which they were laminated since specific gravity differed between a constituent (A) and a constituent (B) is not involved in base phase of a constituent (A). Therefore, if curing treatment is presented simultaneously, a silicone complex which consists of hardening base phase (silicon gel) of a constituent (A) and a hardening

surface protection layer (a silicone elastomer or silicone resin) of a constituent (B) provided in the surface by an adhesion condition will be obtained.

[0018]As for a difference of specific gravity of a silicone composition (A) and a silicone composition (B), it is preferred that it is 0.05 or more, and it is preferred that it is 0.2-0.5 further. A method in particular of establishing such specific gravity difference is not restricted to a silicone composition (A) and a silicone composition (B). A method of illustration can be taken to the following.

[0019]According to the 1st mode of this invention, a bulking agent that specific gravity becomes large from a constituent (B) is added by constituent (A). According to the 2nd mode, a bulking agent and/or an organic solvent in which specific gravity becomes small from a constituent (A) are added by constituent (B). Of course, it is also possible to add a bulking agent which increases specific gravity to a constituent (A) as the 3rd mode, and to add a bulking agent which decreases specific gravity to a constituent (B), and a solvent. Hereafter, order is explained later on.

[0020]A silicone composition (A) generates a gel hardened material, i.e., silicon gel, by hardening. Silicon gel is specified to JIS K-6301 and means a silicone hardened material whose rubber hardness measured with A type spring-loaded hardness scale is 0. [as used herein] Gel means that a silicone hardened material is in a state of such silicon gel. Generally, this silicon gel has the three-dimensional network structure with a low degree of cross linking, and it changes with stress, and vibration is absorbed or it shows mobility. On the other hand, an elastomer means a hardened material in which said rubber hardness is size and has the hardness of a range below 100 from 0, and said rubber hardness with resin. A hardened material which has hardness of 100 or more is meant.

[0021](a) Organopolysiloxane (a) of organopolysiloxane \*\*, It averages among all the organic groups combined with a silicon atom contained in one molecule. More than 0.025 mol % is an alkenyl group, And an alkenyl group which has the viscosity at 25 \*\* in the range of 10 - 100,000 cP and which was combined with a silicon atom in one molecule is averaged. It has 0.1-2 pieces. Silicon gel will be hard to be obtained if there are few rates of an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule than average 0.025-mol %. As this alkenyl group, a vinyl group, an allyl group, an isopropenyl group, a cyclohexenyl group, etc. are mentioned. (a) Since it reacts easily according to that composition of an ingredient is easy, and many kinds of catalysts, a vinyl group is preferred.

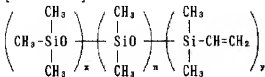
[0022]As organic groups other than an alkenyl group combined with a silicon atom, A methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, Aralkyl groups, such as alkyl groups, such as an octyl group and a decyl group, benzyl, and 2-phenylethyl group, Aryl groups, such as a phenyl group, a tolyl group, and a naphthyl group, or a chloromethyl group which replaced some or all of a hydrogen atom of these bases with a halogen atom etc., 1-10 carbon atoms, such as a 3,3,3-trifluoropropyl group, -- a monovalent hydrocarbon radical of unsubstituted [with 1-8 carbon atoms] or substitution is mentioned preferably. Since composition being easy, and the heat resistance of silicon gel obtained and a physical property are excellent, a methyl group is preferred.

[0023]viscosity at 25 \*\* of this organopolysiloxane -- 10 - 100,000 cP -- it is 100 - 10,000cP preferably. If viscosity is lower than 10cP, workability will fall that it is easy to flow through a silicone composition (A) obtained, and physical properties, such as a loss tangent etc. of a hardened material further obtained by hardening of this constituent (A), will become dissatisfied. On the other hand, if viscosity is higher than 100,000cP, the workability of a constituent obtained will worsen.

[0024]In order for a hardened material produced by making harden a silicone composition (a) to become a gel thing, (a) an average of 0.025 among all the organic groups combined with a silicon atom which contains an ingredient in one molecule -- it is required to be what more than mol % is an alkenyl group, and has 0.1-2 average alkenyl groups combined with a silicon atom in one molecule. Straight chain shape or branched state may be sufficient as molecular structure of this organopolysiloxane (a). A form of these mixtures may be sufficient.

[0025]What is expressed with the following general formula as organopolysiloxane of (a) ingredient which was stated above, for example is mentioned.

[Formula 1]



[Here, x+y=2 and n are positive integers in the positive number of 0<=x<=1.9 and 0.1<=y<=2.]

[0026](b) An ORGANOHAIDOROJIEN PORISHIROKISANORUGANO hydrogen polysiloxane (b), It must react to the alkenyl group in aforementioned (a) ingredient, silicon gel must be formed, and the hydrogen atom combined with a silicon atom must exist in [ 1-50 ] one molecule. Such a hydrogen atom may be combined with the end of the ORGANO hydrogen polysiloxane chain, or which intermediate silicon atom.

[0027]What was illustrated as bases other than the alkenyl group combined with the silicon atom of organopolysiloxane of the aforementioned (a) as univalent atoms or bases other than the hydrogen atom combined with the silicon atom of the ORGANO hydrogen polysiloxane (b), and the same thing can be illustrated. (b) Since an ingredient's being easily compoundable and the heat resistance of silicon gel obtained become the more outstanding thing, a methyl group is preferred.

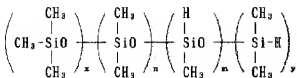
[0028]The ORGANO hydrogen polysiloxane (b), It is necessary to have organopolysiloxane (a) and compatibility, and, for that purpose, organic groups other than an alkenyl group in organopolysiloxane (a), There should be just few silicon atoms which have an organic group of a different kind in (b) ingredient, and it is preferred that the above-mentioned silicon atom is usually less than 10 mol % to all the silicon atoms in the ORGANO hydrogen polysiloxane (b).

[0029](b) Loadings of an ingredient, A hydrogen atom combined with a silicon atom in (b) ingredient to one alkenyl group combined with a silicon atom in the (a) ingredient. 0.3-2.0 They are an individual and the quantity which will be 0.5-1.5 pieces preferably. From 0.3 piece, into silicon gel obtained as it is a small quantity, an unreacted alkenyl group remains and the heat resistance of a hardened material falls. There is a possibility of foaming in the case of hardening of a constituent (A) obtained as there are more quantity than 2.0 pieces.

[0030]In order for a hardened material which a silicone composition (A) generates by hardening to become a gel thing The (b) ingredient needs to be what has the hydrogen atom combined with a silicon atom in [ 1-50 ] one molecule., Although viscosity in particular is not restricted Since easy to compound the (b) ingredient and workability are good, in 25 \*\*, it is preferred that it is the range of 10 - 1,000 cP.,

[0031]What is expressed with the following general formula as an ORGANO hydrogen polysiloxane of (b) ingredient which was stated above, for example is mentioned.

[Formula 2]



[Here, x and y are the number of x>=0, y>=0, and x+y=2, and m and n are integers with which 0<=m<=250, 0<=n<=50, and 0<=m+n<=250 are filled.]

[0032](c) Addition reaction catalyst addition reaction catalyst, Alkenyl group combined with the silicon atom in the (a) ingredient, What kind of catalyst conventionally known as what promotes an addition reaction (hydrosilylation reaction) with the hydrogen atom combined with the silicon atom in the (b) ingredient may be sufficient. It is used by the platinum metal system catalyst and Usually, for example, chloroplatinic acid, denaturing alcohol chloroplatinic acid, Platinum system catalysts, such as a complex of chloroplatinic acid and a vinyl siloxane, and a chloroplatinic acid-2-ethylhexanol solution, and tetrakis (triphenyl phosphine) Palladium system catalysts, such as a mixture of palladium, palladium black, and triphenyl phosphine, a rhodium catalyst, etc. are mentioned. A chloroplatinic acid-2-ethylhexanol solution is especially preferred.

[0033]What is called a catalyst amount may be sufficient as loadings of these catalysts. usually as opposed to the total quantity of the (a) ingredient and (b) ingredient -- it is the range of 0.1 - 100 ppm (catalyst metallic element conversion).

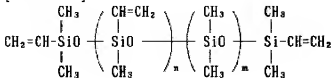
[0034]When the 1st mode of the specific-gravity-adjustment agent above raises specific gravity of a constituent (A) from specific gravity of a constituent (B), a filler is added as a specific-gravity-adjustment agent. Specific gravity this filler. 1.5 or more are preferred and two to about seven are more preferred. As such a filler, inorganic bulking agents, such as fumed silica, sedimentation nature silica, grinding quartz, diatomite, iron oxide, titanium oxide, and calcium carbonate, manganese carbonate, carbon black, alumina, silicon carbide, nitrogen silicon, etc. are mentioned, for example.

[0035]loadings of a bulking agent are chosen so that a silicone composition (A) may have the above-mentioned specific gravity difference to a silicone composition (B), but the range of 10-100 weight section may usually be sufficient as them per total quantity 100 weight section of (A) ingredient and (B) ingredient.

[0036]By hardening, a silicone composition (B) constituent (B) generates a silicone elastomer or silicone resin. When a constituent (B) becomes the shape of an elastomer, or the shape of resin by hardening Organopolysiloxane of the (d) ingredient, More than average 0.05 mol % is an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule, And it is a hydrogen atom which it is required in one molecule to be what has two or more alkenyl groups combined with a silicon atom, and combined the ORGANO hydrogen polysiloxane (e) with a silicon atom in one molecule 5-100 It is required to be what \*\*\*\*.

[0037]As organopolysiloxane (d) provided with such conditions, what is expressed with the following general formula is mentioned.

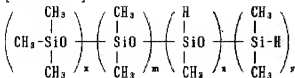
[Formula 3]



[ -- here --n[ m and ] 0 or more integer]

[0038]As an ORGANO hydrogen polysiloxane (e), what is expressed with the following general formula is mentioned.

[Formula 4]



[Here, x and y are the number of  $x \geq 0$ ,  $y \geq 0$ , and  $x+y=2$ , and m and n are  $0 \leq m$  and an integer of  $3 \leq n \leq 100$ .]

[0039]It is possible by changing the presentation of a hardenability silicone composition (B), and the amount of this constituent (B) used to adjust the hardness and thickness of a surface layer. It is concrete, The amount of alkenyl groups in the (d) ingredient, Thickness of a surface layer can be thickened by being able to raise the hardness of a hardened material and increasing the amount of the constituent (B) used by increasing the quantity of the silicon atom absorbed water matter atom (Si-H group) in the (e) ingredient.

[0040]When making specific gravity of a constituent (B) lower than specific gravity of a constituent (A) by the 2nd mode of the specific-gravity-adjustment agent above, a filler and/or \*\*\*\*\* are added by constituent (B) as a specific-gravity-adjustment agent. this filler and solvent -- specific gravity [ ] -- 0.9 or less are preferred -- 0.5-0.8 grade is more preferred. Specifically, foam, such as solvents, such as halogenation hydrocarbon system solvents, such as hydrocarbon system solvents, such as toluene, xylene, and hexane, chlorofluocarbon, and a carbon tetrachloride, silicone foam, and urethane foam, hollow silica powder, hollow phenol resin powder, etc. are mentioned. Especially, especially hollow silica powder is preferred.

[0041]Loadings of a bulking agent or a solvent are chosen so that a silicone composition (B) may have the above-mentioned specific gravity difference to a silicone composition (A), but the range of 10 per total quantity 100 weight section of (a) ingredient and (b) ingredient - 50 weight section may usually be sufficient as them. A solvent is useful to cast a constituent (A) and to form a uniform surface layer on a phase which also lowers viscosity of a constituent (B) and it not only reduces specific gravity of a constituent (B), but consists of constituents (A).

[0042]In other combination drug silicone compositions (A) and silicone compositions (B), an additive agent in which adding conventionally to a hardenability silicone composition besides the aforementioned ingredient is known may be added further if needed. For example, they are colorant, such as flame retarders, such as reaction controlling agents, such as an acetylenealcohol compound, manganese carbonate, and carbon black, a color, and paints, heat-resistant stabilizer, oilproof stabilizer, etc. In order to raise the vibration absorption of a hardened material produced by hardening, an organic polymer bulking agent containing a low boiling compound may be blended with an inside if needed. However, a constituent (A) needs to add these arbitrary additive agents so that specific gravity may not injure relatively a relation that it is small, from a constituent (B).

[0043]In order to provide a phase which consists of a process silicone composition (A) which provides a phase which consists of silicone compositions (A), there is [ which is poured / which is sprayed / which applies this constituent (a) / on the inside of a mold of predetermined shape made into the purpose, or a support surface / in ] a method of making it extruding [ to eject ], being dropped, fall, etc.

[0044]It applies [ which applies a silicone composition (B) ] to the surface of a phase which consists of a

silicone composition (A) which is the process above which provides a layer which consists of silicone compositions (B), and was made and provided by pouring [ to spray ] in, ejecting, extruding, etc., and a surface layer of a constituent (B) is provided in it. Since specific gravity of a constituent (B) is larger than specific gravity of a constituent (A), a surface layer is not mutually mixed with base phase.

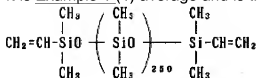
[0045] Although that by which an unhardened surface layer was laminated by unhardened base phase by a process of the curing process above is obtained, curing treatment is simultaneously presented with these as one. A silicone complex which consists of a surface layer which consists of base phase, an elastomer, or resin which consists of silicon gels by this can be obtained. As conditions for hardening, the usual curing conditions may be sufficient, for example, it is the temperature about 60 - 150 \*\*, and is 30-180. Heat-treatment about a part may be sufficient.

[0046] Since a layer of a silicone elastomer or resin whose internal silicon gel phase is a surface layer as a stress relaxation phase functions effectively as a surface protection layer, a silicone complex obtained by a method of use this invention is useful as coating of various kinds of electrical and electric equipment and the surface of electronic parts or a semiconductor device. A silicone complex of this invention can be fabricated on any surface of substrates, such as various kinds of plastics, metal, and glass.

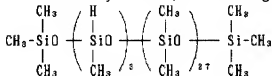
[0047]

[Example] In the following statements, viscosity shows the value at 25 \*\*.

It is Example 1 (1) average and is the following formula. : [Formula 5]

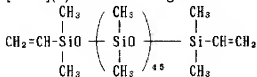


Dimethylpolysiloxane 100 weight section which it is come out and expressed (vinyl group content 0.4-mol %), and is viscosity 1000cP, the following formula : [Formula 6]

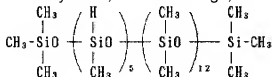


[0048] It was come out and expressed, ORGANO hydrogen polysiloxane 5 weight section of viscosity 28cP, 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions, and crystalline grinding silica 35 weight section were taught to the mixer, it mixed uniformly, and the silicone composition (A-1) was obtained.

[0049] (2) It is an average and is the following formula. : [Formula 7]



It is come out and expressed (vinyl group content 2.1-mol %), is dimethylpolysiloxane 100 weight section of viscosity 60cP, and an average, and is the following formula. : [Formula 8]

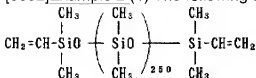




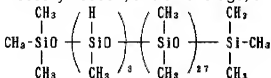
[0050] It is expressed [ (however, this formula is what expressed the number of constitutional units for convenience, and these units may be arranged at random), and ], ORGANO hydrogen polysiloxane 7 weight section of viscosity 10cP and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions were taught to the mixer, were mixed uniformly, and the silicone composition (B-1) was obtained.

[0051] (3) 20 g of the aforementioned silicone compositions (A-1) were poured into an aluminum petri dish 5 cm in diameter, it was made uniform thickness, and, subsequently to the surface, 2 g of silicone compositions (B-1) were laminated uniformly. When this laminated constituent was heated at the temperature of 60 °C for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and gel base phase which has cold resistance was obtained.

[0052] Example 2 (1) The following structural formula : [Formula 9]

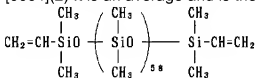


It is come out and expressed (0.40 mol of vinyl group content %), is organopolysiloxane 100 weight section of viscosity 1000cP, and an average, and is the following formula. : [Formula 10]

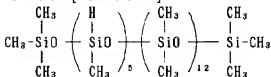


[0053] It is expressed [ (however, the constitutional unit in a formula is arranged at random) and ], ORGANO hydrogen polysiloxane 5 weight section of viscosity 28cP, 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions, and crystalline grinding silica 35 weight section were taught to the mixer, it mixed uniformly, and the silicone composition (A-2) was obtained.

[0054] (2) It is an average and is the following formula. : [Formula 11]



It is come out and expressed and is viscosity. Organopolysiloxane 100 weight section of 100cP, the following formula : [Formula 12]



[0055] It is expressed [ (however, the constitutional unit is arranged at random among the formula), and ], After teaching ORGANO hydrogen polysiloxane 7 weight section of viscosity 10cP, and 5 ppm (platinum conversion) of chloroplatinic acid-2-ethylhexanol solutions to the mixer and mixing uniformly, the obtained mixture was diluted twice using toluene and the silicone composition (B-2) was obtained.

[0056] (3) The 20 g aforementioned silicone composition (A-2) was poured into an aluminum petri dish 5 cm in

diameter, it was considered as uniform thickness, and, subsequently to the surface, 4 g of silicone compositions (B-2) were laminated uniformly. When this laminated constituent was heated at the temperature of 60 °C for 2 hours, the transparent silicone complex which consists of a surface layer of the shape of an elastomer without adhesiveness and gel base phase which has cold resistance was obtained.

[0057]

[Effect of the Invention]According to this invention, it has high hardness, high intensity, high solvent resistance, etc., and the silicone complex which serves as hardness and a surface layer with easy regulation of thickness from the base phase which has low hardness, low intensity, high solvent resistance, adhesiveness, etc. can be easily manufactured by one curing treatment.

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CLAIMS

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[Claim(s)]

[Claim 1]It is a manufacturing method of a silicone complex which consists of a surface layer which consists of a silicone elastomer or silicone resin formed in base phase which consists of silicon gels, and its surface, (a) inside 0.025 [ an average of ] of all the organic groups combined with a silicon atom contained in one molecule -- more than mol % being an alkenyl group, and, . And viscosity at 25 \*\* is in the range of 10 - 100,000 cP. An alkenyl group combined with a silicon atom is averaged in one molecule. Organopolysiloxane which it has 0.1-2 pieces, (b) It is the ORGANO hydrogen polysiloxane which has the hydrogen atom combined with a silicon atom in [ 1-50 ] one molecule, Per [ which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a) ] alkenyl group 0.3-2.0 What is blended so that it may become an individual, And a process of providing a phase which consists of a hardenability silicone composition (A) containing the (c) addition reaction catalyst which hardens and generates silicon gel, Next, it is (d) on a phase of said constituent (A). More than average 0.05 mol % is an alkenyl group among all the organic groups combined with a silicon atom contained in one molecule, . And viscosity at 25 \*\* is in the range of 10 - 100,000 cP. Organopolysiloxane which has two or more alkenyl groups combined with a silicon atom in one molecule, (e) It is a hydrogen atom combined with a silicon atom in one molecule 5-100 It is the ORGANO hydrogen polysiloxane which \*\*\*\*, Per [ which the number of these hydrogen atoms combined with a silicon atom in said organopolysiloxane (a) ] alkenyl group 0.3-2.0 What is blended so that it may become an individual, And contain the (f) addition reaction catalyst and specific gravity is smaller than said constituent (A), A manufacturing method which has the process of presenting hardening with a layer which consists of a process of providing a layer which consists of a hardenability silicone composition (B) which hardens and turns into a hardened material of the shape of an elastomer, or the shape of resin, a phase which consists of said constituent (A), and a constituent (B) simultaneously.

[Claim 2]A way it is the method according to claim 1, and said constituent (A) is a thing containing a bulking agent that specific gravity becomes large from said constituent (B).

[Claim 3]A way it is the method according to claim 1 or 2, and said constituent (B) is what contains further a bulking agent and/or an organic solvent in which specific gravity becomes small from said constituent (A).

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[Translation done.]